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14. ABSTRACT  This project targets mathematical and computational underpinnings of a predictive capability for high performance, nano-composite (HPNC) materials. The overall goal is two-fold: 1 <sup>st</sup> we develop theory, models, and numerical algorithms for the HPNC processing pipeline, starting with composite information, through flow processing, and finally to multi-functional property characterization; 2 <sup>nd</sup> we implement these mathematical tools to predict measurable and significant features of HPNC materials.				
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## HIGH PERFORMANCE MACROMOLECULAR MATERIALS

AFOSR F49620-03-1-0098  
AFOSR FA9550-06-1-0063

Final Report  
Progress Report

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**Overview** This project targets mathematical and computational underpinnings of a predictive capability for high performance, nano-composite (HPNC) materials. The overall goal is two-fold: 1<sup>st</sup> we develop theory, models, and numerical algorithms for the HPNC processing pipeline, starting with composite information, through flow processing, and finally to multi-functional property characterization; 2<sup>nd</sup> we implement these mathematical tools to predict measurable and significant features of HPNC materials.

The materials we consider are nano-rods and nano-clays in aqueous and polymeric solvents, which are technology targets for multi-functional properties ranging from electrical, thermal, mechanical, and barrier (low permeability of liquids and low diffusivity of gases). We have made progress in the theory and models for flow processing and effective properties, in the analysis, numerical algorithms and simulations of the models, and in scientific understanding of nano-composite materials. A current high profile project relates to nano-particle contacts, which are significant even at extremely low volume fractions (~1%) due to the extreme aspect ratios of nano-particles. The role of contacts, and percolation thresholds, across composition and processing space is critical to property characterization, and is now experimentally implicated as a dominant effect. The PI is collaborating with Dr. Richard Vaia, AFRL/MLBP, Wright-Patterson AFB, Dayton, OH, on this critical issue; Vaia has directed the PI's focus on effective properties, providing both the experimental evidence for causal effects as well as feedback on predictions coming out of our modeling group.

During and as a consequence of flow processing, the nano-particle ensemble is theoretically described in terms of an orientational distribution function (PDF). In extension-dominated flow, such as fiber processing, features of the PDF have been shown by the authors in previous contracts to be predictable and controllable. In film and mold-filling processing, anomalous dynamical responses of the nano-ensemble and generation of spatial gradient morphology have been the subject of intense experimental, theoretical and computational scrutiny for the past two decades. The theory has simply not been available to make predictions of relevance to technology, and in particular, the mathematical community has only recently engaged these challenges. Forest and collaborators have made significant progress on a controlled understanding of model experiments in which the flow is shear-dominated, representative of film and mold processing, in the coupling of extensional and shear flow components, in the role of confinement through molecular anchoring on solid boundaries which leads to gradients in

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the PDF of the nano-ensemble and significant flow feedback (nonlinear shear bands). We have recently generalized the Doi-Hess-Marrucci-Greco models and predictions to incorporate an imposed magnetic field, which may serve as a controller during processing to shape the PDF and rid the ensemble of deleterious orientational shear-dominated features. We have further made significant advances in mapping flow-induced PDFs of the nano-ensemble to effective properties, both for homogenized averaging properties and recently for nano-particle cluster and percolation dominated properties. All of these projects are linked together, serving as a basis for an eventual control strategy that bridges composition, processing, effective properties, and performance evaluation.

Specifically, we build theory, models, and simulations that provide the link from control parameters: composition (viscous solution or polymer matrix plus nano-elements), processing flow type and rates, and confinement conditions (device lengthscales as well as solid boundaries); to multi-functional material properties.

A significant obstruction in the production pipeline is control over anisotropy and heterogeneity of performance properties in nano-composites, which arise on lengthscales between the molecular and device scales. These property features are clearly a consequence of the orientational morphology of the anisotropic nano-inclusions, which are typically long rod-like or thin platelet molecules with aspect ratios between 20 and 2000. At present, these anisotropic ensembles and spatial morphologies of the nano-inclusions, and subsequently of the effective properties of materials, are viewed as problematic. However, these features are only possible in soft matter materials, and offer potential keys to novel materials. We currently have theory, modeling, and simulation predictions with a host of the relevant chemical composition and processing physics built in. Other physics and chemistry are not on solid theoretical foundation yet, such as non-uniform mixing of the nano-elements and a polymeric solvent rather than a viscous liquid, we have made progress in incorporating them into the framework.

In our approach, the design and control pipeline is divided into a sequence of fundamental theory and computation problems:

- Molecular potentials and process controls determine the micron-scale, physical structures due to molecular orientational distributions generated in films and molds, as well as stored elastic stresses in these viscoelastic composites. The mathematical theory and models, analytical solution methods, and numerical simulation tools for these orientational anisotropy and structure properties and associated stored stresses are central components of our research over the funding period of **F49620-03-1-0098**. The extension of the numerical algorithms for processing to full 2D and 3D physical space, and their implementation and understanding into a coherent picture, are key projects in the renewal **FA9550-06-1-0063**.
- Once the micron-scale molecular morphology is characterized, either as a numerical database, analytical scaling properties, or an experimentally determined dataset, the next challenge to mathematics and computation is the determination

of effective material performance properties. Since there are millions of molecules in a cubic micron, clearly one must develop scale-up methods based on the molecular orientational distribution, molecule properties, the solvent or matrix properties, and the geometry of the material (film thickness, mold shape). Here we have teamed with Robert Lipton, Louisiana State University, to marry the results of composite homogenization theory with our molecular structure morphology results. This gives the effective anisotropic composite property tensor, parametrized by composition and processing parameters, which we have now published in 4 major articles supported by **F49620-03-1-0098**. Through interactions with R. Vaia, we have identified HPNC systems where percolation and contacts among nano-particles are dominant in properties, as opposed to homogenized averaging. This feature is a high priority of our current efforts in the renewal contract **FA9550-06-1-0063**.

- The last stage in the pipeline is a direct solve for the performance features of the film, using the mechanical, thermal, electric, piezoelectric, permeability property tensor(s) of the nano-composite, which are the variable, anisotropic coefficients in an elliptic, second- or fourth-order operator, together with realistic boundary conditions the materials are exposed to during performance conditions. We have only begun to run simulations for this purpose.
- Finally, a control wrapper is necessary that measures performance properties, evaluates them based on a cost functional which penalizes departure from desired properties, and then gives feedback to which composition and processing parameters can minimize cost and thereby achieve desirable performance features. This capability is realistic within the next 3-5 years, and is the central focus of the sub-contract **FA9550-06-C-0017** with Nonlinear Control Strategies.

#### Acknowledgment/Disclaimer

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#### Personnel Supported in part by **F49620-03-1-0098** and **FA9550-06-1-0063**

Eric Choate	Graduate student, UNC (degree expected May, 2007)
Xiaoyu Zheng	Graduate student, UNC (degree expected May, 2006)
Lingxing Yao	Graduate student, UNC (degree expected May, 2007)
Ruhai Zhou	Postdoc, UNC, 2002-2005
Zhenlu Cui	Postdoc, UNC, 2005-present
Hong Zhou	Consultant, UC Santa Cruz and Naval Postgraduate School
Max Gunzburger	Consultant, Florida State University

## **Publications since 2003 (project period for F49620-03-1-0098)**

1. Full-tensor alignment criteria for sheared nematic polymers (with R. Zhou, Q. Wang), *J. Rheology* (1), 105-128 (2003).
2. Monodomain response of finite-aspect-ratio macromolecules in shear and related linear flows, (with Q. Wang), *Rheologica Acta*, 20-46 (2003).
3. An integrable model for stable:unstable wave coupling phenomena, (with O. Wright), *Physica D* , 173-189 (2003).
4. Computational observation of a weakly compressible mixing barrier in idealized anelastic fluid equations, (with R. McLaughlin and H. Zhou), *Physics of Fluids* 15(10), 2872-2885 (2003).
5. Thermal expansion models of viscous fluids based on limits of free energy, (with S. Bechtel, F. Rooney, and Q. Wang), *Physics of Fluids* 15(9), 2681-2693 (2003).
6. The weak shear kinetic phase diagram for nematic polymers, (with Q. Wang, R. Zhou), *Rheologica Acta*, Volume 43, Number 1, 17-37 (2004).
7. Internal Constraint Theories for the Thermal Expansion of Viscous Fluids, (with S.E. Bechtel, F.J. Rooney), *International Journal of Engineering Science*, Vol. 42, 43-64 (2004).
8. Structure scaling properties of confined nematic polymers in plane Couette cells: the weak flow limit, (with Q. Wang, H. Zhou, R. Zhou), *J. Rheology*, Volume 48(1), 175-192, January/February (2004).
9. Scaling behavior of kinetic orientational distributions for dilute nematic polymers in weak shear, (with Q. Wang, R. Zhou), *JNNFM* Volume 116, Issue 2-3, 183-204 (2004).
10. A kinetic theory for solutions of nonhomogeneous nematic liquid crystalline polymers with density variations, (with Q. Wang, R. Zhou), *Journal of Fluids Engineering*, Volume 126, 180-188 (2004).
11. Monodomain response of arbitrary aspect ratio nematic polymers in general linear planar flows, (with Q. Wang, R. Zhou, E. Choate), *JNNFM*, Volume 118(1), 17-31 (2004).
12. Kinetic theories and mesoscopic models for solutions of nonhomogeneous liquid crystal polymers, (with C. Calderer, Q. Wang), *JNNFM*, Volume 120(1), 69-78 (2004).
13. Likelihood & expected-time statistics of monodomain attractors in sheared discotic and rod-like nematic polymers, (with Zheng, X., Zhou, R., Wang, Q.), *Rheol. Acta*, Volume 43(1), 17-37 (2004).
14. The flow-phase diagram of Doi theory for sheared nematic polymers, II: finite shear rates, (with Zhou, R., Wang, Q.), *Rheol. Acta*, Vol. 44(1), 80-93 (2004).
15. Chaotic boundaries of nematic polymers in mixed shear and extensional flows, (with R. Zhou, Q. Wang), *Physical Review Letters*, Volume 93(8), 088301, August (2004).
16. Exact scaling laws for electrical conductivity properties of nematic polymer nano-composite monodomains, (with X. Zheng, R. Lipton, R. Zhou, Q. Wang), *Advanced Functional Materials*, Volume 15(4), 627-638, April (2005).
17. Kinetic structure simulations of nematic polymers in plane Couette cells, I: The algorithm and benchmarks, (with R. Zhou, Q. Wang), *SIAM Multiscale Modeling and Simulation*, Volume 3(4), 853-870 (2005).

18. Extension-enhanced conductivity of liquid crystalline polymer nano-composites, (with H. Zhou, X. Zheng, Q. Wang, R. Lipton), *Macromolecular Symposia*, Volume 28, 81-85 (2005).
19. A numerical study of unsteady, thermal, glass fiber drawing processes, (with H. Zhou), *Communications in Mathematical Sciences*, Volume 3(1), March (2005).
20. Connections between stability, convexity of internal energy, and the second law for compressible Newtonian fluids, (with S.E. Bechtel, F. Rooney, and Q. Wang), *ASME J. Applied Mechanics*, (2005).
21. Anisotropy and dynamic ranges in effective properties of sheared nematic polymer nano-composites, (with X. Zheng, R. Zhou, Q. Wang, R. Lipton), *Advanced Functional Materials*, Vol. 15 (12), 2029-2035 (cover graphic for December issue) (2005).
22. Hydrodynamic theories for mixtures of polymers and rodlike liquid crystalline polymers, (with Q. Wang), *Physical Review E*, Volume 72, 041805: 1-17 (2005).
23. Anisotropy and heterogeneity of nematic polymer nano-composite film properties, (with R. Zhou, Q. Wang, X. Zheng, R. Lipton), *Institute for Mathematics and Its Applications*, Volume 141, **Modeling of Soft Matter**, 85-98 (2005).
24. A new proof on uniaxial equilibria of a 3-dimensional Smoluchowski equation, (with H. Zhou, H. Wang, and Q. Wang), *Nonlinearity*, Volume 18, 2815-2825 (2005).
25. Kinetic structure simulations of nematic polymers in plane Couette cells, II: In-plane structure transitions, (with R. Zhou, Q. Wang), *SIAM Multiscale Modeling and Simulation*, Volume 4(4), 1280-1304 (2005).
26. Alignment and rheo-oscillator criteria for sheared nematic polymer films in the monolayer limit, (with J. Lee, R. Zhou), *Discrete and Continuous Dynamical Systems*, Volume 6, 339-356 (2006).
27. Anchoring distortions coupled with plane Couette & Poiseuille flows of nematic polymers in viscous solvents: morphology in molecular orientation, stress & flow, *Discrete and Continuous Dynamical Systems*, Volume 6, 407-425 (2006).
28. On weak plane Couette and Poiseuille flows of rigid rod and platelet ensembles, (with Z. Cui, Q. Wang, H. Zhou), *SIAM J. Applied Math*, in press (2006).
29. A classical problem revisited: Rheology of nematic polymer monodomains in small amplitude oscillatory shear, (with E. Choate), *Rheologica Acta*, to appear (2006).
30. Nematic liquids in weak capillary Poiseuille flow: structure scaling laws and effective conductivity implications, (with H. Zhou), *Int. J. Numerical Analysis & Modeling*, to appear (2006).

#### **Submitted refereed research articles and web-posted preprints**

31. Spatial coherence, rheological chaotic dynamics, and hydrodynamic feedback of nematic polymers in plate-driven shear, (with R. Zhou, Q. Wang), *Phys. Rev. Lett.*, submitted July, 2005, in revision.
32. Anchoring-induced structure transitions of flowing nematic polymers in plane Couette cells, (with H. Zhou, Q. Wang), *JNNFM*, to be submitted, April, 2006.
33. A correspondence principle between Stokes flows of viscous and viscoelastic fluids, (with I. Klapper, K. Xu), *JNNFM*, to be submitted, March, 2006.
34. Monodomain dynamics for rigid rod & platelet suspensions in strongly coupled

coplanar linear flow and magnetic fields, (with Q. Wang, R. Zhou), *J. Rheology*, submitted, February, 2006.

35. Characterization of stable kinetic equilibria of rigid, dipolar rod ensembles for coupled dipole-dipole and excluded-volume potentials, (with H. Zhou, H. Wang, Q. Wang), *Nonlinearity*, submitted, January, 2006.

36. Autoregressive & maximum likelihood methods for microrheological characterization of viscoelastic media, (with J. Fricks, L. Yao, T. Elston), *J. Rheology*, to be submitted, March, 2006.

37. Extension of the Ferry shear wave model for viscoelastic characterization to finite depth, wave reflection, and nonlinearity, (with S. Mitran, B. Lindley, L. Yao), *JNNFM*, to be submitted, March, 2006.

38. Nano-rod suspension flows: a 2D Smoluchowski-Navier-Stokes solver, (with R. Zhou, Q. Wang), *Int. J. Numerical Analysis & Modeling*, submitted, March (2006).

#### **Honors & Awards Received by the PI**

Grant Dahlstrom Distinguished Professor of Mathematics, UNC, 2003

#### **AFRL Point of Contact**

Richard Vaia, AFRL/MLBP, Bldg 654, WPAFB, OH, Phone 937-255-9184.

#### **Presentations (since 2003 that acknowledge AFOSR support)**

1. 2003, "Laminar flows of nematic polymers: issues critical to high-performance materials", *University of Colorado-Boulder, Applied Mathematics Colloquium, Boulder, CO, Jan. 18*
2. 2003, "Mathematics of high-performance materials", *AMS Special Session on the Mathematics of Materials, Rob Lipton, Organizer, Baton Rouge, LA, March 14*
3. 2003, "Laminar flows of nematic polymers: passing between kinetic and mesoscopic scale models", *Workshop on Multiscale Theory and Computation in Nano Materials, Wright-Patterson AFB, Dayton, OH, March 29*
4. 2003, "What's math got to do with nano-materials?", *UNC-Chapel Hill Mathematics Graduate Visions Seminar, April 14*
5. 2003, "Laminar flows of nematic polymers: high-performance materials in the making", *Carnegie Mellon University, Mathematics Department and Center for Nonlinear Analysis, Pittsburgh, PA, May 1*
6. 2003, "The dynamics of nematic polymers in laminar flows: a zoo of bifurcations associated with bulk molecular phase transitions", *SIAM Dynamical Systems Annual Meeting, Snowbird, UT, May 28*
7. 2003, "Theory, modeling, and simulation of soft-matter nano-composites", *NASA URETI meeting, Langley, Virginia, August 20*

8. 2003, "Laminar flows of nematic polymers: high performance materials in the making", UNC Applied Mathematics Seminar, September 19
9. 2003, "Laminar flows of nematic polymers: high performance materials in the making", Florida State University, Mathematics Colloquium, September 22
10. 2003, "Structure phenomena in confined flows of nematic polymers", Society of Rheology Annual Meeting, Pittsburgh, October 15
11. 2003, "Multiscale challenges in soft matter materials", American Mathematical Society Regional Meeting, University of North Carolina, October 20
12. 2004, "Nano-composite material properties: homogenization over flow-induced orientational distributions", Gordon Research Conference Poster on Colloidal, Macromolecular & Polyelectrolyte Solutions, February 3
13. 2004, "Bridging computational algorithms for control & design of multi-functional nano-composite properties", Conference on Emerging Technologies in Computation and Partial Differential Equations, CSIT, Florida State University, March 10
14. 2004, "Polymer Nano-Composite Properties: A Roadmap for Design & Control", American Mathematical Society Regional Meeting, Tallahassee, FL, March 12
15. 2004, "A Control and Design Roadmap for Multi-functional, Polymer Nano-Composite Properties", NJIT Applied Mathematics Colloquium, Newark, NJ, April 23
16. 2004, "Connecting Molecular Orientation and Structure to Nano-Composite Properties", Air Force Office of Scientific Research Annual Meeting of Contractors and Grantees, Wright Patterson AFB, Dayton, OH, June 14
17. 2004, "High Order, Reliable Computation of Flow Structure in Nematic Polymers", Numerical Methods in Forming of Polymeric Materials, Columbus, OH, June 15
18. 2004, "Connecting Molecular Orientation and Structure to Nano-Composite Properties", Second International Times of Polymers Meeting, Ischia, Italy, June 21
19. 2004, "Multifunctional, multiscale property patterning of high performance nano-composite materials", SIAM Annual Meeting, Portland, OR, July 15
20. 2004, "Multiscale Mathematics of Complex Fluids and Soft Matter", Department of Energy Workshop on the Multiscale Mathematics Initiative, Denver, CO, July 20

21. 2004, "What's Math Got to Do with High-Performance Nano-Composites?", College of Charleston, Southeastern Applied Mathematics Workshop, Charleston, SC, September 17
22. 2004, "What's Applied and Computational Math Got to Do with High-Performance Nano-Composites?", Princeton University Program in Applied & Computational Mathematics Colloquium, Princeton, NJ, October 5
23. 2004, "A modeling thrust: Modeling the nano-composite pipeline for nano-composite materials", NASA BiMat URETI annual review, Langley, VA, October 8
24. 2004, "Toward predictive capability in medicine", UNC Vice Chancellor's Workshop on Nanomedicine, Chapel Hill, NC, October 12
25. 2004, "Hydrodynamics & the isotropic-nematic phase transition: consequences for nano-composite material properties", Duke Center for Nonlinear and Complex Systems, Colloquium, Durham, NC, October 26
26. 2005, "Modeling and simulation of nematic polymer film flows and their effective material properties", North Carolina State University, colloquium in Mechanical & Aerospace Engineering, Raleigh, NC, January 18
27. 2005, "Modeling the pipeline of high performance, nano-composite materials and effective properties, I and II", NSF Institute for Mathematics and Its Applications, Workshop on Composites: Where Mathematics Meets Industry, Minneapolis, MN, February 8
28. 2005, "Nematic polymer materials: from hydrodynamics to effective properties", U. Wisconsin at Madison, Rheology Research Center, "Mohs Lectures by Placon" Lecture Series, February 11
29. 2005, "Anisotropy and heterogeneity of nematic polymer nano-composite film properties", American Chemical Society Annual Meeting, Polymer Nanocomposites Symposium, San Diego, CA, March 15
30. 2005, "Nano-composite materials", UNC Chapel Hill, Department of Mathematics Colloquium, Chapel Hill, NC, April 7
31. 2005, "Predictive Multi-scale Tools for Nano-Composite Processing & Effective Multifunctional Properties", NASA BiMat Annual Meeting, May 6.
32. 2005, "Mathematical challenges in nano-composite materials", Center for Scientific Computation & Applied Mathematics, U. Maryland, May 11.
33. 2005, "Modeling of Nano-Composite Materials: from Flow Processing to

*Effective Properties", Air Force Office of Scientific Research Annual Meeting, August 29.*

34. 2005, "A Suite of Models and Algorithms for Nano-Composite Materials", *Iowa State Computational Methods and Applied PDE Workshop, November 5.*
35. 2005, *Flows and effective properties of rod and platelet nano-composites, Penn State Applied Mathematics & Analysis Seminar, November 15.*
36. 2005, *Modeling of Nano-Composite Materials: from Flow Processing to Effective Properties AFOSR-AFRL/ML-Industry Technical Interchange, Dayton, OH, December 5.*
37. 2005, *Nanorod & Nanoplatelet Composite Properties versus Processing, NASA BIMat Program Review, Langley, VA, December 9.*

### **Technology Assists**

The Forest group has established the only proof-of-principle results on HPNC materials whereby effective properties are directly linked to composition and processing phases. These results have led to contacts from two technology companies, Moldflow Corporation and Fujiyama Polymer Research, both of whom are interested in applications of our property characterization tools.

### **Key Accomplishments**

The Forest group has several successes which are recognized as significant accomplishments in referee reports, communications from journal editors, and in our property bifurcation diagram making the cover graphic of the December, 2005 issue of Advanced Functional Materials. These successes are summarized as follows:

**Processing:** Phase diagrams of the stable ensemble probability distribution functions (PDFs) of nematic polymers across processing parameter space. Results include PDF databases and scaling properties versus geometry and volume fraction of the nano-particles, flow type and flow rate, confinement geometry, driving conditions (drag versus pressure) and boundary anchoring conditions. New theory and models, new algorithms, and new identification of phenomena and transition behavior have been published and recognized.

**Property Characterization:** Volume averaged conductivity and mechanical properties versus processing-induced PDFs, and percolation phenomena based on processing-induced PDFs, of nematic polymer nano-composites. Results include effective property tensors in the form of databases and scaling properties versus geometry and volume fraction of the nano-particles, flow type and flow rate, confinement geometry, driving conditions (drag versus pressure) and boundary anchoring conditions. Extensions of existing homogenization theory have been development, along with new algorithms, and

identification of new phenomena and transition behavior have been either published or will be submitted in the upcoming month.

Graphics and powerpoint slides, with annotations, of these accomplishments have been forwarded to the cognizant program officer, Dr. Arje Nachman, and can be modified and updated upon request.